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Uncertainty quantification in non-equilibrium molecular dynamics simulations of thermal transport

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ABSTRACT

Bulk thermal conductivity estimates based on predictions from non-equilibrium molecular dynamics (NEMD) using the so-called direct method are known to be severely under-predicted since finite simulation length-scales are unable to mimic bulk transport. Moreover, subjecting the system to a temperature gradient by means of thermostatting tends to impact phonon transport adversely. Additionally, NEMD predictions are tightly coupled with the choice of the inter-atomic potential and the underlying values associated with its parameters. In the case of silicon (Si), nominal estimates of the Stillinger-Weber (SW) potential parameters are largely based on a constrained regression approach aimed at agreement with experimental data while ensuring structural stability. However, this approach has its shortcomings and it may not be ideal to use the same set of parameters to study a wide variety of Si-based systems subjected to different thermodynamic conditions. In this study, NEMD simulations are performed on a Si bar to investigate the impact of bar-length, and the applied temperature gradient on the discrepancy between predictions and the available measurement for bulk thermal conductivity at 300 K by constructing statistical response surfaces at different temperatures. The approach helps quantify the discrepancy, observed to be largely dependent on the system-size, with minimal computational effort. A computationally efficient approach based on derivative-based sensitivity measures to construct a reduced-order polynomial chaos surrogate for NEMD predictions is also presented. The surrogate is used to perform parametric sensitivity analysis, forward propagation of the uncertainty, and calibration of the important SW potential parameters in a Bayesian setting. It is found that only two (out of seven) parameters contribute significantly to the uncertainty in bulk thermal conductivity estimates for Si.

1. Introduction

Classical molecular dynamics (MD) is commonly used to study thermal transport by means of phonons in material systems comprising non-metallic elements such as carbon, silicon, and germanium [1]. A major objective for many such studies is the estimation of bulk thermal conductivity of the system. One of the most commonly used approaches, regarded as the direct method [2–10], is a non-equilibrium technique that involves the application of a heat flux or a temperature gradient by means of thermostatting, across the system. The corresponding steady-state temperature gradient in the former or the heat exchange between the two thermostats in the latter, is used to estimate the bulk thermal conductivity (at a given length or size) using Fourier's law. However, when the simulation domain is comparable to or smaller than the mean free path, thermal conductivity estimates from the direct method depends on the distance between the two thermostats, due to significant contribution of boundary scattering. Hence, to estimate the bulk thermal conductivity, computations are performed for a range of system lengths and the inverse of thermal conductivity is plotted against the inverse of length. The y-intercept of a straight line fit to the observed trend is considered as the bulk thermal conductivity estimate.

Although widely used, the direct method is known to severely under-predict the bulk thermal conductivity compared to experimental measurements [11,12]. This is primarily due to length scales used in the simulation that are several orders of magnitude smaller than those used in an experiment. As a result, the sample length is much smaller than the bulk phonon mean free path leading to the so-called ballistic transport of the phonons. The mean free path of such phonon modes is limited to the system size that reduces their contribution to thermal transport. Moreover, the introduction of thermostats typically reduces the correlation

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between vibrations of different atoms potentially reducing the thermal conductivity further [13]. The average temperature gradient experienced by the system could thus be different from the simulation input and is a potential source of uncertainty. Estimation of thermal conductivity using the direct method is therefore impacted by the choice of system size and potentially due to fluctuations in the temperature gradient experienced by the system due to thermostatting.

Predictions of non-equilibrium molecular dynamics (NEMD) simulations are also dependent on the choice of the inter-atomic potential as well as values associated with individual parameters of a given potential. For instance, in the case of crystalline Si, the Stillinger-Weber (SW) inter-atomic potential is widely used. However, as discussed by Stillinger and Weber in [14], their proposed nominal values of individual parameters were based on a limited search in a 7D parameter space while ensuring structural stability and reasonable agreement with experiments. It is therefore likely that these nominal estimates for individual parameter values in the SW potential may not yield accurate results for a wide variety of Si-based systems and warrant further investigation to study the impact of underlying uncertainties on MD predictions. Along these lines, a recent study by Wang et al. performed uncertainty quantification of thermal conductivities from equilibrium molecular dynamics simulations [15]. Rizzi et al. focused on the effect of uncertainties associated with the force-field parameters on bulk water properties using MD simulations [16]. Marepalli et al. in [17] considered a stochastic model for thermal conductivity to account for inherent noise in MD simulations, and study its impact on spatial temperature distribution during heat conduction. Jacobson et al. in [18] implemented an uncertainty quantification framework to optimize a coarse-grained model for predicting the properties of monoatomic water. While these are significant contributions, it is only recently that researchers have started accounting for the presence of uncertainties in MD predictions in a systematic manner. There is a definite need for additional efforts aimed at efficiency and accuracy to enable uncertainty analysis in MD simulations for a wide range of applications.

In the present work, we focus our efforts on uncertainty analysis in the predictions of NEMD simulations for phonon transport using a silicon bar. An overview of the set-up for the simulations is provided in Section 2. As discussed earlier, predictions from NEMD exhibit large discrepancies with experimental observations depending upon system size and potentially due to fluctuations in the applied temperature gradient. Additionally, the thermal conductivity estimates are tightly coupled with parameter values associated with the inter-atomic potential. Hence, we set out to accomplish multiple objectives through this research effort: First, we construct response surfaces in order to characterize the dependence of discrepancy in thermal conductivity estimates (between MD simulations and experiments) on system size, and applied temperature gradient (Section 3). Second, we perform sensitivity analysis to study the impact of SW potential parameter values on uncertainty in the predictions (Section 4). Third, we exploit our findings from sensitivity analysis to construct a reduced order surrogate for uncertainty analysis (Section 5). Fourth, we illustrate the calibration of important parameters in a Bayesian setting to evaluate their posterior distributions (Section 6). Construction of the response surfaces, parametric sensitivity analysis, and Bayesian calibration can all be computationally challenging endeavors especially in situations involving compute-intensive simulations as in the case of NEMD. We therefore employ polynomial chaos (PC) surrogates [19,20] using non-intrusive spectral approaches [21] to reduce the computational effort pertaining to the aforementioned objectives. Moreover, since the construction of a PC surrogate itself can be expensive, we demonstrate a novel approach in Section 4 that implements derivative-based sensitivity measures [22] to reduce the dimensionality of the surrogate a priori while ensuring reasonable predictive accuracy.

2. NEMD set-up

In this work, non-equilibrium molecular dynamics (NEMD) simulations are performed using the LAMMPS [23] software package. Essentially, a temperature gradient is applied by means of Langevin thermostats located at $L/4$ and $3L/4$ in a silicon bar of length, L as shown using a schematic in Fig. 1.

The set of inputs to LAMMPS is provided below in Table 1. Note that specific values for the length of the bar and the applied temperature gradient are not provided since we investigate thermal conductivity trends for a range of values of the two parameters as discussed later in Section 3. A careful analysis focused on minimizing temperature fluctuations during different stages of the simulation was performed to optimize for the choice of height and width of the bar as well as the duration of the simulation.

The NEMD simulation has three stages associated with it as illustrated below in the flow diagram. In the first stage, the NVT ensemble equilibrates the system to a specified bulk temperature, i.e., the temperature at which thermal conductivity is to be estimated. In the second stage, the NVE ensemble equilibrates the thermostats at their respective temperatures. It is followed by another NVE ensemble that captures the trajectory of individual

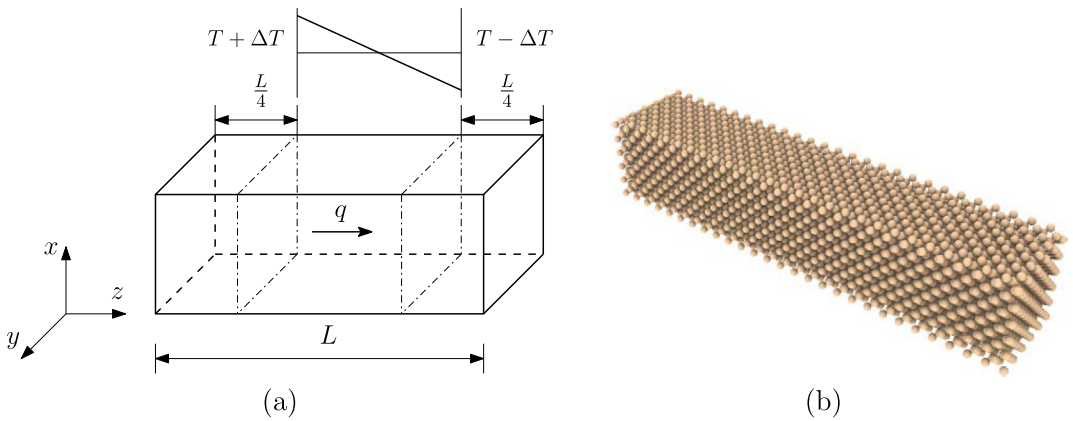

Fig. 1. (a) Schematic illustration of the set-up for evaluating thermal conductivity of Si using NEMD. (b) Arrangement of Si atoms prior to the application of temperature gradient.